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Detection of structural heterogeneity in a hyperquenched silicate glass

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The heterogeneous structure and dynamics in supercooled liquids and their glassy states is an important key to understand the nature of glass and glass transition. Here we study the structural heterogeneity in a ternary silicate glass using hyperquenching–annealing–calorimetry approach [1,2]. We observed two separated sub-T_g relaxation peaks on the calorimetric curve of the hyperquenched CaO–MgO–SiO_2 glass, which have been attributed to two distinct structural domains of higher and lower potential energies, respectively. The higher energy domains in nanoscale are so unstable that they become ordered during hyperquenching. This is verified by the high-resolution transmission electron microscopy (HRTEM) images exhibiting ordered nanodomains in the glass matrix. Moreover, the nanoscale domains are further confirmed by the change of the lower temperature crystallization peak with sub-T_g annealing degree. The pre-ordered domains could lower the activation energy of crystal growth above T_g, and thus they remain stable during the sub-T_g annealing. But the higher temperature crystallization peak varies with the sub-T_g annealing degree, implying that the sub-T_g annealing affects only the lower energy domains, i.e., the disordered glass matrix. In addition, the higher energy domains during relaxation behave as a strong glass phase, whereas the lower energy domains do as a fragile one. It is found that silica crystals form at 1084 K for 4 hours, indicating the existence of the typical strong phase – silica in the supercooled magnesium-calcium silicate. It is also found that both silica and diopside form at 1084 K for 24 hours or at 1283 K for 2 hours, indicating the existence of the typical fragile phase – diopside. Therefore, the coexistence of the two types of local structures in the hyperquenched CaO–MgO–SiO_2 glass is a clear manifestation of the structural heterogeneity in the supercooled liquid.

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[2] Y. F. Zhang, L. N. Hu, S. J. Liu, C. F. Zhu, Y. Z. Yue, Sub-T_g enthalpy relaxation in an extremely unstable oxide glass and its implication for structural heterogeneity, J. Non-Cryst. Solids **381** (2013) 23-28.